

# Atmospheric CO<sub>2</sub> stabilization and ocean acidification

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[1] We use a coupled climate/carbon-cycle model to examine the consequences of stabilizing atmospheric CO<sub>2</sub> at different levels for ocean chemistry. Our simulations show the potential for major damage to at least some ocean ecosystems at atmospheric CO<sub>2</sub> stabilization levels as low as 450 ppm. Before the industrial revolution, more than 98% of corals reefs were surrounded by waters that were >3.5 times saturated with respect to their skeleton materials (aragonite). If atmospheric CO<sub>2</sub> is stabilized at 450 ppm only 8% of existing coral reefs will be surrounded by water with this saturation level. Also at this CO<sub>2</sub> level 7% of the ocean South of 60°S will become undersaturated with respect to aragonite, and parts of the high latitude ocean will experience a decrease in pH by more than 0.2 units. Results presented here provide an independent and additional basis for choosing targets of atmospheric CO<sub>2</sub> stabilization levels.  
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## 1. Introduction

[2] Article 2 of the United Nations Framework Convention on Climate Change (UNFCCC) calls for the stabilization of atmospheric greenhouse gas concentrations at levels that “prevent dangerous anthropogenic interference with the climate system”, but the UNFCCC does not define “dangerous interference”. Usually, studies on the choice of stabilization target for the concentration of atmospheric CO<sub>2</sub>, the most important anthropogenic greenhouse gas, is based on the consideration of its radiative effects, such as the magnitude and rate of temperature change [Caldeira *et al.*, 2003; Den Elzen and Meinshausen, 2006; Harvey, 2007]. However, increasing atmospheric CO<sub>2</sub> concentrations not only interfere with the climate system by trapping infrared radiation, but also affect marine ecosystems by perturbing ocean chemistry [Royal Society, 2005].

[3] One consequence of atmospheric CO<sub>2</sub> dissolving into seawater is the formation of carbonic acid (H<sub>2</sub>CO<sub>3</sub>), which releases hydrogen ions (H<sup>+</sup>) and decreases pH, a process referred to as ocean acidification [Caldeira and Wickett, 2003]. Excess hydrogen ions react with carbonate ions (CO<sub>3</sub><sup>2-</sup>) to form bicarbonate ions (HCO<sub>3</sub><sup>-</sup>), reducing [CO<sub>3</sub><sup>2-</sup>] and the ocean’s saturation state with respect to aragonite and calcite, two major forms of calcium carbonate (CaCO<sub>3</sub>). These changes in ocean chemistry could impact marine ecosystems in various ways. A decrease in the saturation state of calcium carbonate can weaken the ability

of calcifying organisms to make calcium carbonate shells [Kleypas *et al.*, 2006], especially for reef-building corals that build their skeleton materials out of aragonite, a more soluble form of CaCO<sub>3</sub> [Gattuso *et al.*, 1998; Leclercq *et al.*, 2000; Langdon and Atkinson, 2005]. As seawater becomes undersaturated with respect to calcium carbonate (saturation state < 1), the shells of calcifying organisms are increasingly prone to dissolution, as has been observed for pteropods which produce shells out of aragonite [Orr *et al.*, 2005]. Furthermore, a decrease in ocean pH can affect the growth of marine organisms by altering the acid-base balance within their cells, the availability of nutrients, and the effects of toxins [Royal Society, 2005].

[4] Here we use a coupled climate/carbon-cycle model to provide information that can help to determine what atmospheric CO<sub>2</sub> stabilization level would constitute “dangerous interference” based on the consideration of changes in ocean chemistry. Compared to previous modeling studies on ocean acidification [Kleypas *et al.*, 1999; Caldeira and Wickett, 2003; Orr *et al.*, 2005; Caldeira and Wickett, 2005], this is the first study presenting geographical distributions of ocean chemistry under different atmospheric CO<sub>2</sub> stabilization scenarios obtained from the simulation of a coupled climate/carbon-cycle model. The results presented here can help in the development of an independent and additional basis for choosing targets of CO<sub>2</sub> stabilization levels and allowable CO<sub>2</sub> emissions.

## 2. Methods

### 2.1. Model

[5] The model we use in this study is the University of Victoria (UVic) Earth System Climate Model version 2.8 [Weaver *et al.*, 2001]. The UVic model is an earth system model of intermediate complexity that consists of a 19-layer general ocean circulation model coupled to an atmosphere, sea-ice, and carbon cycle component. The ocean carbon cycle and carbonate chemistry are represented by an inorganic carbon cycle component based on the protocol of Ocean Carbon Cycle Intercomparison Project (OCMIP, <http://www.ipsl.jussieu.fr/OCMIP>) with an addition of a simple marine biology component.

### 2.2. Simulation Experiment Setup

[6] To examine the consequence of atmospheric CO<sub>2</sub> stabilization concentrations for ocean chemistry we construct a set of CO<sub>2</sub> pathways in which atmospheric CO<sub>2</sub> concentrations are stabilized between 280 and 2000 ppm (Figure S1 of the auxiliary material<sup>1</sup>). CO<sub>2</sub> stabilization pathways considered include stabilization at pre-industrial (280 ppm) and present-day (380 ppm) levels, between 450

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**Table 1.** Percentage area of ocean waters with certain chemistry changes at different atmospheric CO<sub>2</sub> stabilization levels

CO <sub>2</sub>	% Global Ocean	% Ocean south of 60°S		% Coral Reef	
	$\Delta pH > 0.2^a$	$\Omega_{\text{aragonite}} < 1$	$\Omega_{\text{calcite}} < 1$	$\Omega_{\text{aragonite}} > 3.5$	$\Omega_{\text{aragonite}} > 3.25$
280 ppm	0%	0%	0%	98.4%	99.8%
380 ppm	0%	0%	0%	38%	76%
450 ppm	11%	7%	0%	8%	34%
550 ppm	97%	50%	0%	0%	2%
650 ppm	99.8%	86%	5%	0%	0%
750 ppm	100%	95%	10%	0%	0%
1000 ppm	100%	100%	68%	0%	0%

<sup>a</sup> $\Delta pH$  is the decrease in global mean surface ocean pH relative to pre-industrial value of 8.18.

and 1000 ppm at an interval of 50 ppm, and between 1000 and 2000 ppm at an interval of 100 ppm. The stabilization year is 1765 and 2010 for the 280 and 380 ppm pathways, 2100 for the 450 ppm pathway and increases by 25 years for each 50 ppm increase in atmospheric CO<sub>2</sub> stabilization level, to a maximum of year 2375. The UVic model is spun up for 5000 years to a stationary state, and is then integrated from pre-industrial time (year 1765) to year 2500 forced by atmospheric CO<sub>2</sub> concentrations described above. The model-simulated inventory of anthropogenic CO<sub>2</sub> in year 1994 is 111PgC, which is consistent with data-based estimates of  $118 \pm 19$ PgC by *Sabine et al.* [2004] and  $107 \pm 14$  PgC by *Waugh et al.* [2006].

### 2.3. Analysis of Results

[7] To compute ocean chemistry fields we use the chemistry routine from the OCMIP-3 project (<http://www.ipsl.jussieu.fr/OCMIP/phase3>) that uses the model output of dissolved inorganic carbon (DIC), alkalinity (ALK), temperature, and salinity to calculate pH and saturation state of aragonite ( $\Omega_{\text{aragonite}}$ ) and calcite ( $\Omega_{\text{calcite}}$ ). All model fields used here are averaged values over the 50-year period after CO<sub>2</sub> stabilization occurs. In our calculations, we add model-predicted perturbations (relative to year 1994) in dissolved inorganic carbon, alkalinity, temperature, and salinity to the values inferred from observational data [*Key et al.*, 2004]. Model results are used directly where observational data are not available such as in the Arctic Ocean. The resolution of the model (1.8° latitude by 3.6° longitude) does not allow it to resolve coastal regions. Coastal ocean chemistry can vary widely from that of the open ocean [*Feely et al.*, 2008]. However, changes in coastal ocean chemistry should largely track corresponding changes in nearby open ocean waters. To relate open water chemistry to that of coral reefs, we obtain the longitude and latitude location of each coral reef from the Reefbase dataset ([www.reefbase.org](http://www.reefbase.org)) and then interpolate model-predicted nearby open-water values to each reef location. Non-reef building coral communities are excluded in our analysis.

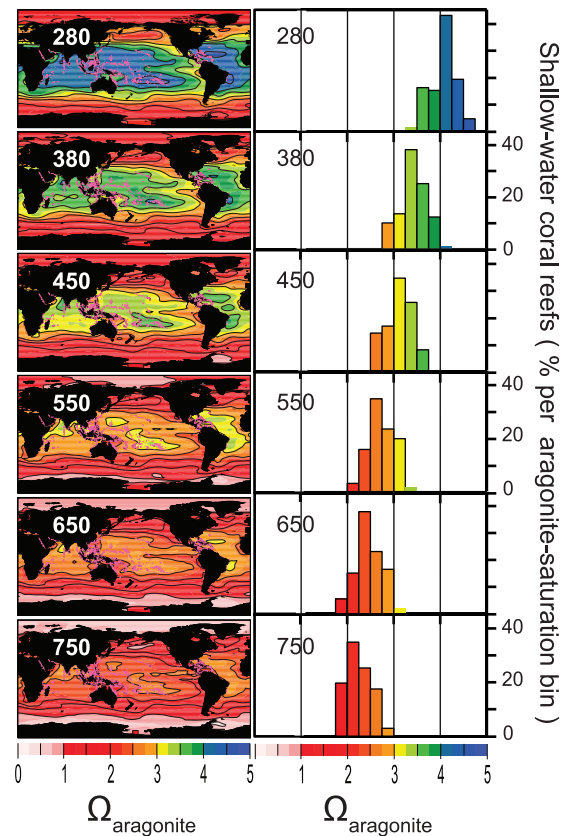
### 3. Results

[8] In the following, we examine the consequence of stabilizing atmospheric CO<sub>2</sub> at different levels for ocean chemistry by focusing on three specific aspects (1) how chemical conditions (e.g., aragonite saturation state) of seawater surrounding existing coral reefs will change under various CO<sub>2</sub> stabilization concentrations, (2) what CO<sub>2</sub> stabilization levels are required to avoid calcite and/or aragonite undersaturation in different parts of the ocean,

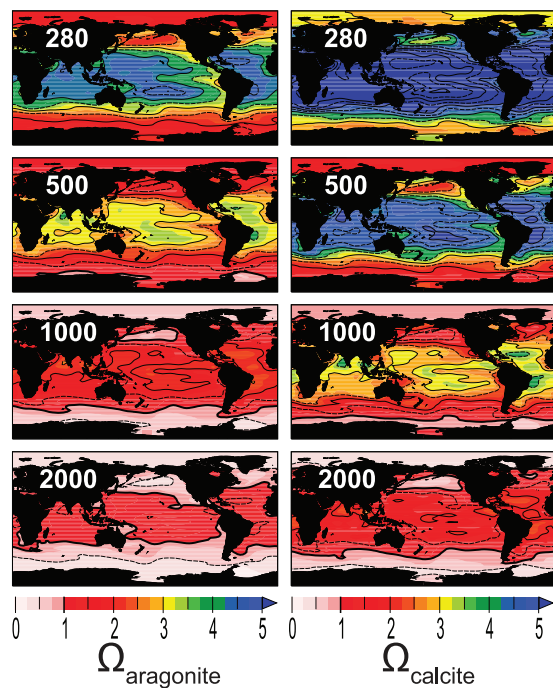
and (3) how ocean pH will change at different CO<sub>2</sub> stabilization levels.

#### 3.1. Aragonite Saturation State and Coral Reefs

[9] Here we examine near-surface aragonite saturation state under various CO<sub>2</sub> stabilization levels and relate it to



**Figure 1.** Aragonite saturation state and coral reefs. (left) Maps of model-predicted aragonite saturation states at different atmospheric CO<sub>2</sub> stabilization concentrations (ppm) plotted over existing shallow-water coral reef locations (shown as magenta dots). (right) Percentage distribution of modern day coral reefs at each aragonite saturation bin under different atmospheric CO<sub>2</sub> stabilization concentrations. Aragonite saturation value at each reef location is interpolated from nearby open ocean values simulated by the model. Results are obtained by adding model-predicted perturbations in geochemical fields to modern observations, except for the Arctic Ocean where results are directly from model simulations as a result of the lack of observations.



**Figure 2.** Saturation state of aragonite and calcite. Saturation state of aragonite ( $\Omega_{\text{aragonite}}$ ) and calcite ( $\Omega_{\text{calcite}}$ ) for different atmospheric CO<sub>2</sub> stabilization levels (ppm). By 500 ppm, aragonite will dissolve in parts of the near-surface Southern Ocean; by 1000 ppm, calcite will dissolve in most of the high latitude ocean. Results are obtained by adding model-predicted perturbations in geochemical fields to modern observations, except for the Arctic Ocean where results are directly from model simulations as a result of the lack of observations.

the chemical condition of existing shallow water coral reefs (Table 1 and Figure 1). Higher atmospheric CO<sub>2</sub> stabilization levels rapidly decrease the aragonite saturation state of open waters surrounding existing coral reefs (auxiliary material). Our analysis show that before the industrial revolution when atmospheric CO<sub>2</sub> level was about 280 ppm, 98.4% of coral reefs were found near open ocean waters with an aragonite saturation state above 3.5. With atmospheric CO<sub>2</sub> stabilized at higher levels waters surrounding coral reefs with such aragonite saturation state disappear rapidly. Even if atmospheric CO<sub>2</sub> could be stabilized at the current level of 380 ppm, 62% coral reefs would be surrounded by waters that are less saturated than 3.5. With a CO<sub>2</sub> stabilization of 450 ppm only 8% of coral reefs would be surrounded by open ocean waters with aragonite saturation state above 3.5, and with a stabilization level of 550 ppm no existing coral reefs would be near such waters. The loss of ocean water with high aragonite saturation surrounding coral reefs has the potential to weaken their calcification ability and raises questions about the long-term stability of carbonate reef structures.

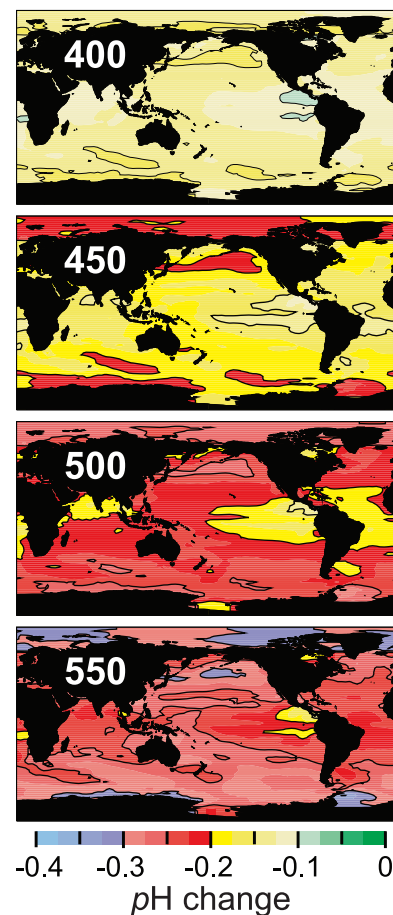
[10] In addition to aragonite a decrease in calcite saturation state (Figure 2) would affect the calcification of marine calcifiers such as coccolithophores. It was widely accepted that calcification of coccolithophores will decrease with a decrease in calcite saturation [e.g., Riebesell *et al.*, 2000; Delille *et al.*, 2005], but a recent study suggest a more

complicated picture [Iglesias-Rodriguez *et al.*, 2008]. Observations of communities living near undersea sources of geologic CO<sub>2</sub> support the inference that high CO<sub>2</sub> levels will favor non-calcifying organisms over calcifying organisms [Hall-Spencer *et al.*, 2008].

### 3.2. Undersaturation of Aragonite and Calcite

[11] What CO<sub>2</sub> stabilization level is needed to avoid widespread dissolution of calcite and aragonite minerals (i.e., saturation state <1) in the near-surface ocean? In preindustrial times, aragonite saturation ranged from 1.4 in cold polar waters to 4.7 in warm tropical waters; calcite saturation ranged between 2.2 and 7.0 (Figure 2). Therefore, with increasing atmospheric CO<sub>2</sub> concentrations, high latitude ocean would be the first to become undersaturated with respect to calcium carbonate.

[12] Our results show that even at a CO<sub>2</sub> stabilization level as low as 450 ppm, parts of the Southern Ocean become undersaturated with respect to aragonite (7% of the ocean area south of 60°S). At 550 ppm, half of the ocean south of 60°S becomes undersaturated with respect to aragonite. At 750 ppm, 95% of this area becomes under-



**Figure 3.** Ocean pH change. Changes in surface ocean pH relative to pre-industrial values for different atmospheric CO<sub>2</sub> stabilization levels. Results are obtained by adding model-predicted perturbations in geochemical fields to modern observations, except for the Arctic Ocean where results are directly from model simulations as a result of the lack of observations.



saturated (Table 1 and Figure 2). In contrast, at 400 ppm, we predict no aragonite undersaturation in open ocean waters of the Southern Ocean. Parts of the Southern Ocean become undersaturated with respect to calcite when atmospheric CO<sub>2</sub> is stabilized above 600 ppm. At 1000 ppm, the area with calcite undersaturation expands to cover 68% of the ocean south of 60°S (Table 1 and Figure 2). If atmospheric CO<sub>2</sub> is stabilized at 2000 ppm, parts of tropical and subtropical oceans (including the Great Barrier Reef regions) will experience aragonite undersaturation, and almost all waters poleward of 50° will experience calcite undersaturation (Figure 2).

### 3.3. pH Change

[13] Now we look at the consequence of stabilizing atmospheric CO<sub>2</sub> for ocean pH change (Figure 3). Our results show that increasing atmospheric CO<sub>2</sub> concentrations over the past two centuries have caused a 0.1 units decrease in average pH for the global surface ocean (corresponds to a 30% increase in hydrogen ion concentrations), consistent with previous studies [Royal Society, 2005]. At the CO<sub>2</sub> stabilization level of 450 ppm, more than 10% of the surface ocean, including a large portion of the Southern, North Pacific, and Arctic oceans, experiences a pH decrease of more than 0.2 units, in violation of the criteria set forth by the U.S. Environmental Protection Agency [1976] that “for open ocean waters...the pH should not be changed more than 0.2 units from the naturally occurring variation”, and the “guard rail” by the German Advisory Council on Global Change [Schubert *et al.*, 2006]. When atmospheric CO<sub>2</sub> is stabilized at 550 ppm, most surface ocean experiences a pH decrease of more than 0.2 units and the high latitude ocean has a pH decrease of more than 0.3 units.

## 4. Discussion and Conclusions

[14] There are several caveats we need to mention here. The model used in this study does not resolve coastal waters, and thus we characterize the chemistry of open ocean water in proximity to coral reefs. In addition to the influence of open water chemistry, chemistry of waters where coral reef systems reside is affected by local processes such as terrestrial and groundwater input, shallow-water carbonate precipitation/dissolution, and advection of deeper ocean waters onto the continental shelves [e.g., Frankignoulle *et al.*, 1996; Kawahata *et al.*, 2000]. Therefore, the aragonite saturation state of coral reef ecosystems might be different from that of the surrounding open waters. In addition to ocean acidification, many factors affect the fate of coral reefs [Hoegh-Guldberg *et al.*, 2007]. Calcification rate of coral reefs may increase with temperature increase, but this increase is unlikely to continue after a temperature rise of 2°C to 3°C [Kleypas *et al.*, 2005]. On the other hand, high ocean temperatures can cause corals to bleach, and lower aragonite saturation state of seawater could compound this effect. In addition, increased temperature around coral reefs (Figure S2) could interact with lower aragonite saturation state to further affect coral reef calcification [Reynaud *et al.*, 2003]. Nutrient availability, declining water quality, and overfishing will also affect coral reefs. Nevertheless, a rapid decrease in the aragonite saturation state of open water

surrounding coral reefs will exacerbate other forms of global and/or local stresses they might experience in the future. Little is known about the potential adaptation and acclimation of coral reefs to lower aragonite saturation states, but no adaptation has yet been observed. Corals polyps may continue to persist under low aragonite saturation conditions in soft-body forms lacking calcified-skeletons [Fine and Tchernov, 2007], but organisms in this state would not be reef builders.

[15] Our prediction of ocean chemistry change is obtained using a coupled climate/ocean-carbon cycle model that includes the effect of climate change (changes in temperature, ocean circulation, etc.) as a result of increasing atmospheric CO<sub>2</sub> concentrations. Our results are broadly consistent with previous studies that did not include the effect of climate change. For example, using an ocean-only model, Caldeira and Wickett [2005] predicted a decrease in global surface pH of 0.24 and 0.47 units in year 2300 when atmospheric CO<sub>2</sub> reaches 550 and 990 ppm respectively. By comparison, we predict a decrease in pH of 0.25 and 0.48 units at CO<sub>2</sub> stabilization levels of 550 and 1000 ppm. Sensitivity experiments confirm that changes in ocean chemistry are mainly caused by increased CO<sub>2</sub> concentrations. For example, the effect of climate change on ocean chemistry is to somewhat diminish the CO<sub>2</sub>-induced decrease in aragonite saturation state around coral reefs (Figure S3). The minor role of climate change in modifications of ocean chemistry has also been reported in previous studies [Orr *et al.*, 2005; McNeil and Matear, 2007; Cao *et al.*, 2007].

[16] We conclude that ocean chemistry will be significantly perturbed even atmospheric CO<sub>2</sub> can be stabilized at low to modest levels. For example, at a stabilization level of 450 ppm, some parts of the high latitude ocean would become undersaturated with respect to aragonite and experience a decrease in pH by more than 0.2 units. At a stabilization level of 550 ppm, there will be no water left in the open ocean with the kind of chemistry (aragonite saturation levels) experienced by more than 98% of shallow-water coral reefs before the advent of the industrial revolution. These changes to ocean chemistry are likely to adversely affect marine ecosystems. Therefore, preservation of existing marine ecosystems could require a CO<sub>2</sub> stabilization level that is lower than what might be chosen based on climate considerations alone.

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